Master's Thesis

Understanding Long-range Transport of Air Pollutants in East Asia Using a Lagrangian Particle Dispersion Model

Hyuckjae Lee

Department of Urban and Environmental Engineering
(Environmental Science and Engineering)

Graduate School of UNIST

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Hyuckjae Lee

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Approved by

Myong In Lee

Advisor

Myong-In Lee

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Hyuckjae Lee

This certifies that the thesis of Hyuckjae Lee is approved.

12. 5. 2018

Myong-In Lee
Ulsan National Institute of Science and Technology

Sung-Deuk Choi
Ulsan National Institute of Science and Technology

Chang-Keun Song
Ulsan National Institute of Science and Technology

Abstract

Nitrogen dioxide (NO_2) is a tracer of Nitrogen oxides (NO_x) and it has been very used for tracking NO_x. In-situ measurements is very helpful of determining atmospheric concentrations. OMI tropospheric NO_2 column retrieval is also used for estimating NO_x in atmosphere. Even though we have such useful instrument and in-situ measurement, there still remains a limitation of spatio-temporal inhomogeneous which means that it is hard to understand a three-dimensional transport mechanism of atmospheric transport of air pollutants in detail. Trans-boundary anthropogenic pollutants from a local or transported has been becoming more serious issues in context of politics and scientific basis. We employed a lagrangian particle dispersion model to get better understanding of 3-dimensional atmospheric transport mechanism.

At this point, to overcome these limitations, we conduct a multi-year model simulation to further understand the long-range and trans-boundary transport, and to better understand the 3-dimensional transport mechanism, we estimate the factors affecting to the transport and carry out sensitivity tests for each factor respectively through Source-Receptor Relationship (SRR). We initially estimated emissions, meteorological conditions and decay time as major factors, and investigate a phased experiment, to control the unexpected experimental results. As results, the factors originally set have an effect, but they have not impact drastically on transport mechanism. However, we could confirm that the cause was a significant contribution to the wind field change in the synoptic weather condition, and atmospheric transport is mainly dependent on meteorological conditions rather than emissions. Of course, the weather conditions are relatively dominant, but the effect of emissions is not that much of small. In other words, long-range trans-boundary transport in the East Asia are comprised of the complicated interactions particularly.

Key words: FLEXPART, Long-range transport, Trans-boundary transport, Air pollutants, NO_x, East Asia, Source-Receptor Relationships

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Chapter I

Introduction

Various forms of air pollutants have been especially increasing in East Asia in regard to the advanced technology and rapid economic growth during the past several decades, which has caused widespread environmental and health problems. As a result of anthropogenic activities, the awareness of increasing the air pollutants and its impact on the human society has been becoming serious. The long-range transport is an important issue arising the awareness of migrations of trans-boundary air pollutants in East Asia, furthermore, how the migration of air pollutants changes following to climate change in the future is very growing in context of scientific and politic issues. The trans-boundary migration of air pollutant from local emission sources potentially leads to the increase of awareness for environmental concerns on the domestic energy markets and politics (Shah et al.,2000; Guttikunda et al., 2001).

Fig. 1. 1. Schematic explanations of role of major air pollutants.

NO_x is one represent of anthropogenic activities, importantly playing as a major precursor of ozone causing air pollution and climate change which significant damages on human society. Many South Korean and Japanese scientists have reported, based on measurement data from local observations, that Northeast Asia has been highly affected by sulfur dioxide and nitrogen oxide emissions from China (Kim et al., 2012a; Ohara et al., 2007; Streets et al., 2006). Current issue for air quality in East Asia is that increasing air pollutions, especially NO_x emissions from China. It will lead to higher levels of ozone downwind area if the increasing NO_x emissions of ozone precursors from China (Fusco and Logan, 2003).

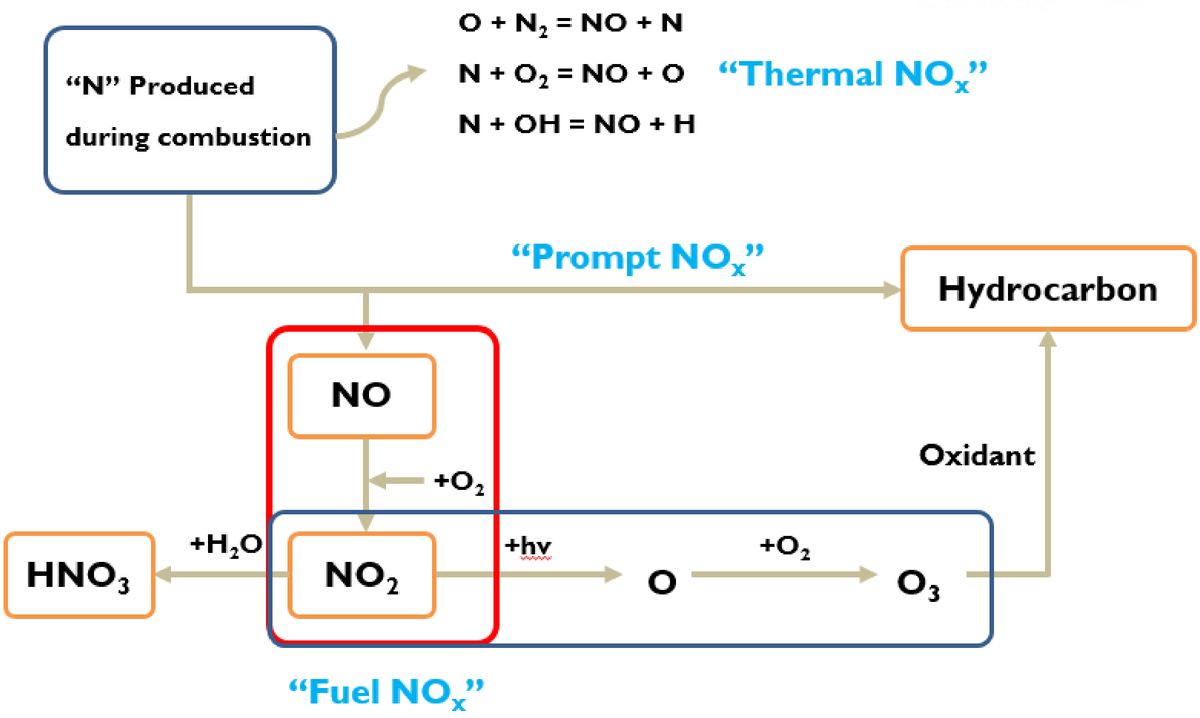

Fig. 1. 2. Schematic diagram of NO_x Formation mechanism.

There are couple of main factors that strongly affect to the long-range transport of emissions over the trans-boundary. Researching for estimation of total emission over the Northeast Asia, where the huge part of total emission in Asia, have been ongoing by many scientists. An accuracy of emission distribution spatially and temporally is important for the air quality modeling framework and forecasts (Ma and van Ardenne, 2004; Eder et al., 2009; Zhang et al., 2012; Struzewska et al., 2016, Ding et al., 2017). In the past few decades, Asian countries have been putting their efforts toward implementing the aim of reducing the emissions below a certain level through mutual co-operations. According to Ding et al (2017), about 10% of emissions reduction over the mainland-China was estimated during Chinese New Year (Winter). Under the scenario of national air pollutants control policies by Chinese government, the air pollutants reduce in the majority of eastern China where the main emission sources (Zhao et al., 2013).

One of representative transport components for describing trans-boundary air pollutants migrations is wind directions and speeds mostly depending on a season. Meteorological features of the winter can be described by Winter monsoon which has the similar characteristics of typical January weather conditions. Winter monsoon is accompanied by Siberian high with cold surges. Winter monsoon determines the local and climate patterns in adjacent East Asia, as well as a considerably effects to the extratropical and tropical planetary-scale circulations (Chang and Lau 1982). According to Chang and Lau (1980), they have reported that cold surges following to winter monsoon can lead to short-term variations of Hadley and Walker circulations and the East Asian jet, which cause the changes to the patterns of wind directions and speeds at adjacent area in East Asia. It can be described that climate change would be able to lead to change weather condition, and influence of the air quality over the adjacent East Asia in winter.

Multi-modeling framework has been conducted by Carmichael et al., (2002), Qu et al., (2016), and Kim et al., (2017b) to get better understanding of various approaches to modeling long-range transport. Even though estimated emissions from emission sources in the local are very closer as far as possible for the real, it is necessary to precede inter-comparison long-range transport modeling studies, due mainly to long-range transport mechanisms of various models are dissimilar in characteristics such as removal process and chemical reactions in different models. Past and ongoing researches for the model simulations has chiefly investigated on seasonal variability of air pollutants in East Asia (Lee et al., 2014; Kim et al., 2013b). However, there is a limit to describe interannual variations of air pollutants with modeling framework. In context of understanding long-range transport mechanism and predicting patterns of air quality in synoptic scale, the modeling framework should be implemented in order to overcome limits of spatial and temporal distributions.

Total amounts of emissions and meteorological conditions are main factors to understand the mechanism for long-range transport of air pollutants, furthermore, the predictions for changes of air quality based on climate change in annual by annual. For each research has been ongoing by many scientists individually, however, researches that analyze the mechanism for trans-boundary long-range transport by interacting synoptic meteorological conditions with emissions through multi-year modeling framework are limited. For doing this, we employ the Source-Receptor Relationships (SRR) analysis which have been widely used for solving the trans-boundary problem related to a potential inter-political conflict and helping for decision making on policies related to environmental. SRR method has examined and investigated for estimation of emissions from various locations in East Asia by Arndt et al. (1998), Huang et al. (1995) and Ichikawa and Fujita (1995), Calori et al. (2001) and the results are very different.

Recently, Cross-country study has been ongoing for this issue (National Institute of Environmental Research, 2012). The research is about long-range trans-boundary pollutants (so called to LTP) in East Asia investigated by experienced researchers from those countries. The purpose of LTP project is to promote understanding of trans-boundary and long-range transport of air pollutants in Northeast Asia through research exchanges and co-operations among the three countries. Consequently, it would be able to provide scientific basis to policy makers to prevent or reduce adverse impacts on the environment in Northeast Asia. As a part of the LTP (Long-range Transboundary Air Pollutants) project, air quality modeling research has been carried out, and studies have been conducted to identify the source-receptor relationship between Korea, China, and Japan (National Institute of Environmental Research, 2012). In case of the year for winter in 2002, it was estimated that 30~40% of nitrates affecting to Korean from China and 45~65 % of estimations on March to South Korea from China were reported. In case of the year of 2006 in winter, they investigated total nitrate (Korea and Japan: HNO_3, NO_3 and PAN / China: NO_x ($NO+NO_2$), HNO_3 and PAN) and 30~60% of contributions from central China to South Korea on March are resulted. The SRR for total amount of estimated nitrate in December was similar to March. Especially, Japan's estimated contribution from source (China) to receptor for oxidized nitrogen depositions in winter is about 60%.

To sum up, we need more understanding of long-range and trans-boundary transport in East Asia, due to limited usability of observations and measurements. Satellite measurement can be represented as a concentration but no pathways of transport and spatial limitations both vertically and horizontally. We are facing these spatiotemporal limitations so that It is hard to understand a three-dimensional transport mechanism of the air pollutants in detail. As a part of multilateral approach, researching for Multi-year model simulation is inquired. In this experiment, we employ the lagrangian FLEXible PARTicle dispersion model (FLEXPART) to investigate the influence of emissions from countries in East Asia to adjacent region. We analyze trans-boundary long-range transport in DJF in which winter monsoon is dominant and focusing on the impact on the Korean peninsula located on the downwind side with analysis of local meteorological conditions as a part of the analyzing the phenomenon. To obtain the knowledge of pollutant transport mechanism in East Asia focusing on winter and to find feasibility of a lagrangian particle dispersion model, this study investigates inter-annually synoptic meteorological conditions in East Asia, standing on the basis of SRR analysis.

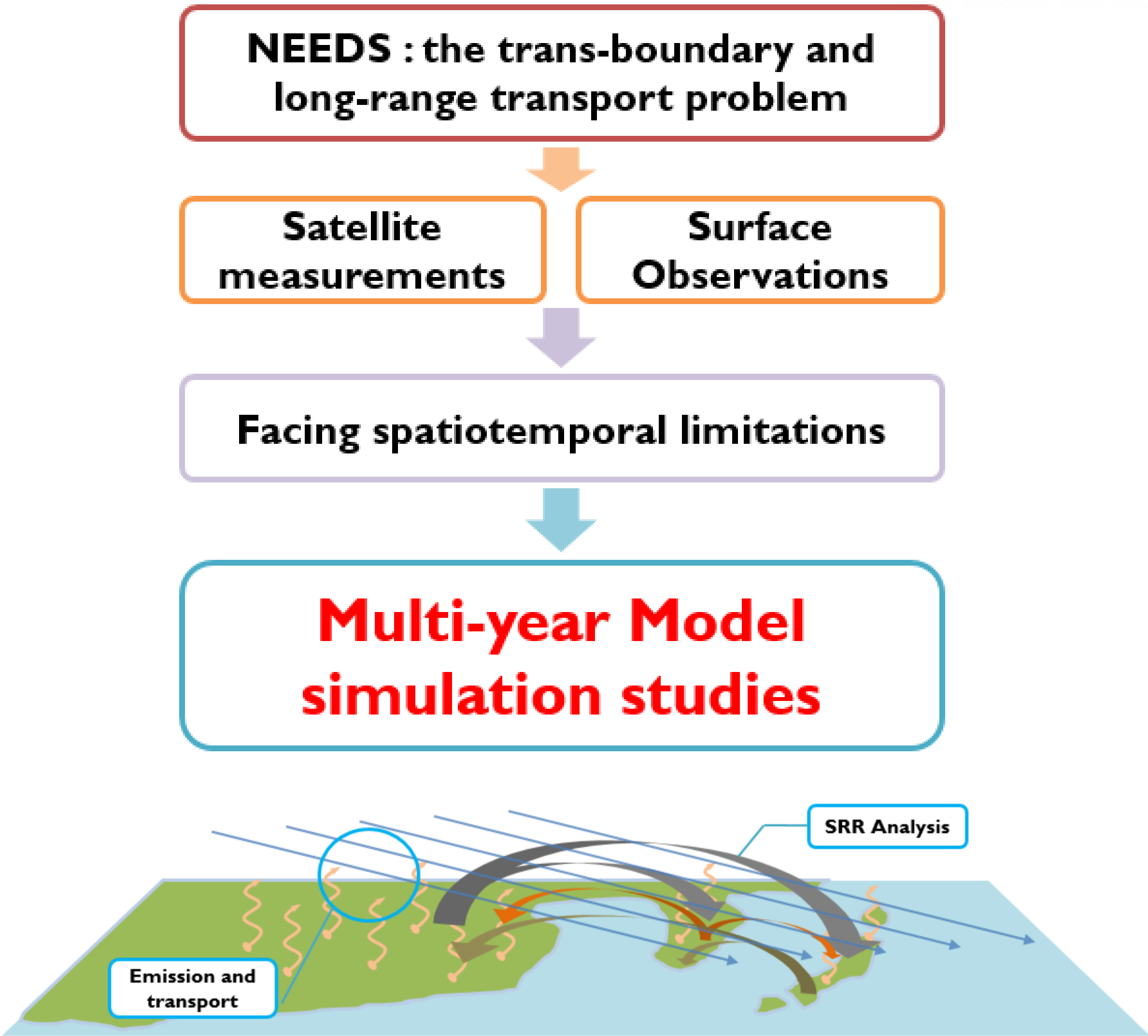

Fig. 1. 3. Schematic for objectives of this study.

Chapter II

Data and Methods

2.1 Model description

FLEXPART is one of representative of the Lagrangian particle dispersion and transport model which widely used for model user community (Stohl et al., 2005). Lagrangian particle models compute trajectories of a large number of particles to demonstrate the transport of particles and trace plume trajectories in the atmosphere. It simulated in various fields of the applications range from dispersion of radioactive materials, anthropogenic and biogenic emissions, and volcanic emissions (Stohl et al., 2007; Brioude et al., 2009; Warneke et al., 2009; Stohl et al., 2011). It also simulates long-range transport and meso-scale transport of particles with calculations of wet and dry deposition and radioactive decay time, released from point, line, area sources (Stohl et al., 2005). In this experiment, we used FLEXPART version of 9.02 for demonstrating the source and receptor areas in East Asia (Lee et al., 2014). This experiment is driven by National Center for Environmental Prediction Climate Forecast System Version 2 (CFSv2) 6 hourly products with horizontal resolution of 0.5° **x** 0.5° and 37 vertical layers, initialized 4 times per day (00, 06, 12, and 18 UTC). Model domain is composed of 0.5° **x** 0.5° horizontal resolution and 16 vertical layers between 0 and 5km. The coverage of model simulations is 90E – 150E and 10N – 60N, over the East Asia. The sectors indicated in figure 2.1 encompass the area of China (black lined), Korea (red dotted lined), and Japan (yellow lined) respectively (also used in Richter et al., 2005 and Lee et al., 2014). Simulations periods are DJF of 2010/11, 2015/16, and 2017/18. NO_2 is a representative anthropogenic emission, as well as a passive anthropogenic NO_x tracer. Target emission in this experiment is NO_2 and 297 area sources of NO_2 at 2° **x** 2° resolution over East Asia. The given set up was assigned to identify source-receptor relationships, covering emissions from either all sources or the sources in a specific area (China, Korea, and Japan) (Lee et al., 2014). In the boundary layer, the lifetime of NO_x is ~6 hour in summer, ~6-12 hour in spring and fall, and ~12 – 20 h in winter due to changes in photolysis rate and water vapor content in the atmosphere (Martin et al., 2003; Lamsal et al., 2010). To avoid absence of chemical conversion in model simulation, we consider decay time for 72 hour as chemical conversion of NO_2, As well as, decay time ranging from 24-hr to 168-hr to investigate sensitivity depending on life-time of NO_2.

2.2 Emission inventory (Comprehensive Regional Emissions inventory for Atmospheric Transport Experiment, CREATE)

Comprehensive Regional Emissions inventory for Atmospheric Transport Experiment (CREATE) emission inventory, which was decided to be used in this study, is an Asian inventory developed through domestic research (Woo et al., 2014). It calculates emissions for the most recent baseline year and presents emissions by local emission systems, including wide target substances and detailed source classifications. CREATE has 54 fuel classes, 201 sub-sectors and 13 pollutants, listed as SO_2, NO_x, CO, NMVOC, NH_3, OC, BC, PM_{10}, $PM_{2.5}$, CO_2, CH_4, N_2O, and Mercury (Woo et al., 2014). CREATE emission inventory integrates extant emission inventories for Korea, China and Japan with emission data-sets, based on the comparisons of emission data determined by sectors and activities (National Institute of Environmental Research, 2013). In addition, it is an emission inventory suitable for simulations and SRR analysis in East Asia, due to regional characteristics of emission sources are considered in detail compared to other global emission inventories. In order to reflect the current state of China, where is the fastest and biggest change in East Asia, it calculates the futuristic coefficients in sectors, regions and substances and create emission inventory (Woo et al., 2014). In order to employ it for model simulations, SMOKE-Asia (v1.3.0) is used to spatial allocation, temporal varying and chemical speciation for CREATE inventory (Woo et al., 2012). There is a couple of previous study using CREATE inventory. Kim et al., (2017a) examined the effects of various emission inventories on predictions of ozone concentrations with model experiments regionally. Basically, they inter-compared to emission inventories they utilized to find sensitivities of each inventory. Lee et al., (2017) also employed CREATE emission inventory to investigate the migrations of various air pollutants using adjoint model in order for understanding the 3-dimensional long-range transport mechanism.

Model	FLEXPART v9.02
Domain	90E-150E / 10N-60N
Resolution	0.5x0.5
Periods	20101201-20110228 / 20151201-20160229 / 20171201 – 20180228 (1011 / 1516 / 1718 DJF)
Input Met. data	National Center for Environmental Prediction Climate Forecast System Version 2(CFSv2) 6-hourly product (0.5x0.5, 37 vertical levels)
Vertical	16 (50, 100, 200, 300, 400, 500, 600, 700, 800, 900, 1000, 1500, 2000, 3000, 4000 and 5000m)
Emission Inventory	**CREATE (2010, 2015), using SMOKE-Asia v1.3.0**
Resolution	2.0 x 2.0 (Actually non-gridded)
Domain	East Asia (Customized)
Target emission	NO_2 (72 hour of decay time), 297 area sources
Temporal resolution	Monthly
Specific.	- Wet scavenging and dry depositions are calculated - Ranging from 24-hr to 168-hr of decay-time - 297 source areas are specified based on CREATE emission inventory, which is used for releases file on FLEXPART

Table 2. 1. Experimental set up for model simulations

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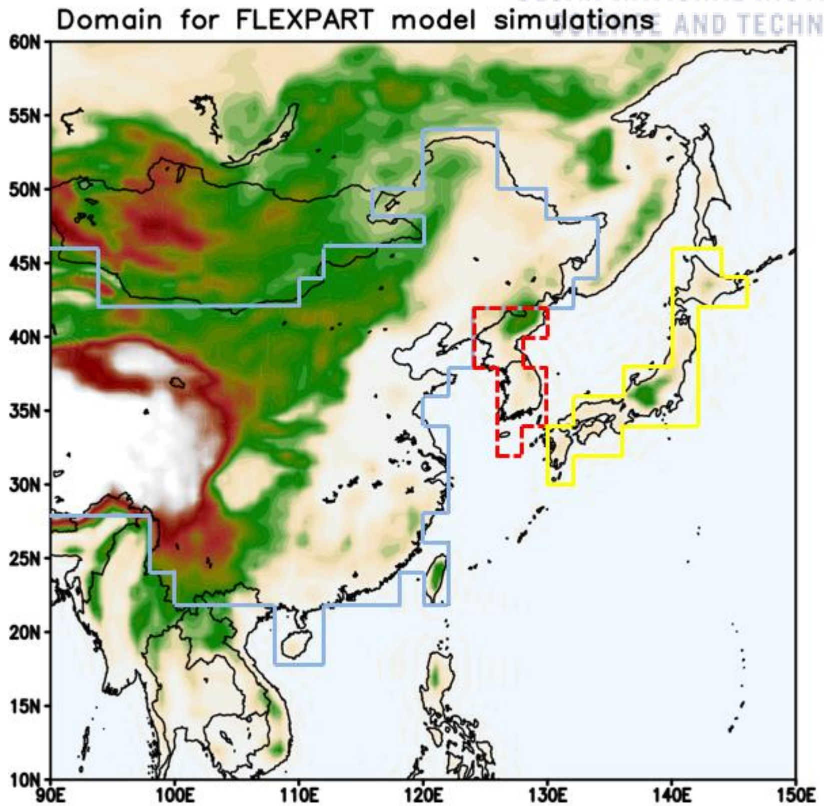


Fig. 2. 1. Domain for FLEXPART model simulations and sectors for SRRs (blue colored for China, red colored for Korea and yellow colored for Japan)

2.3 Observations

Ozone Monitoring Instrument (OMI) onboard Aura satellite launched on 14 July 2004 and has been provided global observations data-set of ozone and key atmospheric air pollutant gases (NO_2 and SO_2) since October 2004 (Krotkov et al., 2015). It provides measurements of solar backscatter, which measures in UV/Visible wavelength ranges (270 to 550nm) to be used tropospheric NO_2 with a spatial resolution up to 13 **x** 24km and daily global coverage (Levelt et al.,2006; Lamsal et al., 2010). The algorithm for producing OMI tropospheric NO_2 data is described by Bucsela et al., (2006, 2008) and Celarier et al., (2008). OMI/Aura NO_2 L3 data is daily global gridded (horizontal resolution of 0.25° x 0.25°**.** Surface measurement data in Korea has been hourly collected by Korea Ministry of Environment, which has 361 monitoring stations. In order for EOFs analysis, we selected major cities in South Kore and put the data into EOF analysis (Seoul, Busan, Dae-gu, In-cheon, Gwang-ju, Daejeon, Ulsan, Chun-cheon, Cheong-ju and Mok-po).

2.4 Source-Receptor Relationship (SRR)

Expression of transport, diffusion and chemical processes taking place in atmosphere is complicated in numerical simulations. One of advantages of air quality model simulation is to be a scientific basis of assess the impacts of emission changes on concentration levels (Clappier et al., 2015). It is often used for simulations how emissions influence the primary chemistries as well as secondary pollutants (Clappier et al., 2015). Specifying the appropriate emission scenarios and simulating model simulations are simple way to determine the concentrations. As we mentioned above, model simulations are not simplified to calculate. Source Receptor Relationships are alternative approach to analyze the transport of air pollutants when uncertain scenarios are faced. Many previous studies for instance have been doing with this approach.

The simple equation we adapted for SRR is shown below:

$$Contribution(i,j) = \frac{From\ source(i,j)}{\sum Receptor\ NO_{2,(i,j)}}$$

For non-linear relationships, the number of coefficients is required following to the degree of non-linearity characterzing the relations, however, linear relationships are demanded only one or no coefficient (Clappier et al., 2015). In FLEXPART, emissions from sources can be calculated separately. Since the sum of the concentrations in the specific coordinate area contains the individual emission information of the region assumed in the model set-up, it is separated into source-receptor relationships. At this point, the FLEXPART model assumes linear relationships because there is no chemical reactions.

Chapter III

Results

3.1 Model validations

As we mentioned earlier, the various attempts of reducing anthropogenic emissions (air pollutants) has been ongoing to improve the air quality in East Asia. As a result of various and active efforts on it, total amounts of air pollutants are getting decreased during past decade as shown in figure 3.1. Figure 3.1 shows that mean OMI tropospheric NO_2 column density from 2010 to 2017 (DJF) and trend of its measurement during that time. This result is seemingly similar to previous study (Souri et al., 2017), which analyzed observations from 2010 to 2014. The figure also includes temporal variations of NO_2 column density during the same period in Seoul, Beijing and shanghai where are the major cities that considerably contribute to the total amount of emissions in East Asia. Mostly, they are decreasing which is matched with the trend. In this experiment, we take the simulation year for 2010, 2015 and 2017 (DJF) which are the base year of emission inventories, as well as, 2011 (DJF) which indicates peak year of OMI-measurement in time-series.

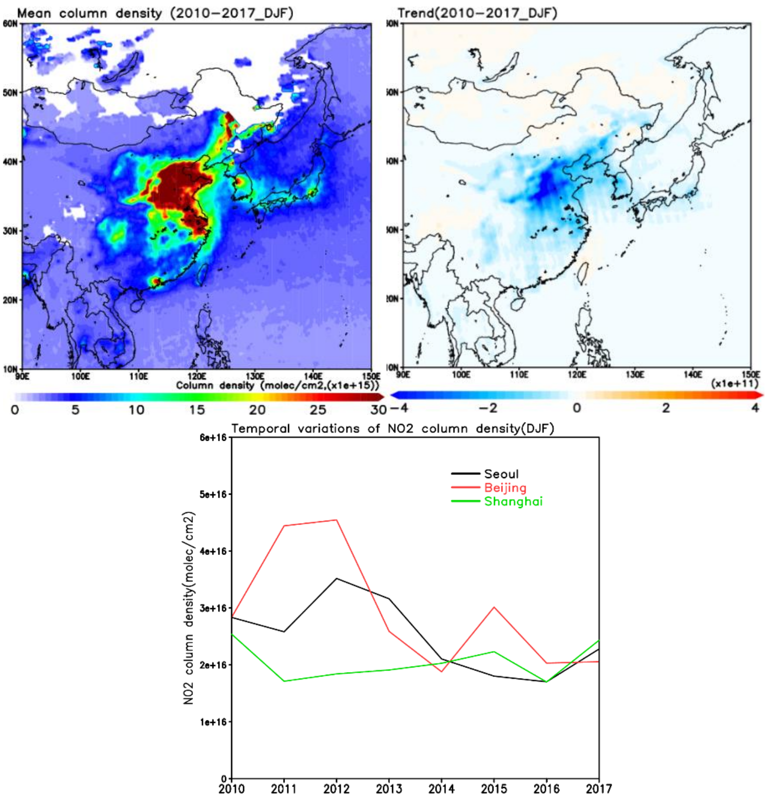


Fig. 3. 1. Spatial distribution of mean tropospheric NO_2 column density (OMI, 2010-2017) in DJF, and trend for NO_2 column density (molec/cm^2, right above) over the East Asia. Temporal variations of Seoul, Beijing and Shanghai where are representative of mega cities and highly polluted areas

Figure 3.2 shows the spatial distributions of mean NO_2 column density in FLEXPART simulations on 2018 January. The date indicated on the figure is selected randomly. Each figure represents (a) simulations results of total sectors, (b) sector for China, (c) sector for Korean and (d) sector for Japan. These were determined based on rough boundary of targeted countries in East Asia as shown in the figure 3.1. The denominator contains the total amount of emissions at target area in (a), numerator contains the total amount of emissions at source area valued in (b), (c) and (d). At a glance, migrations

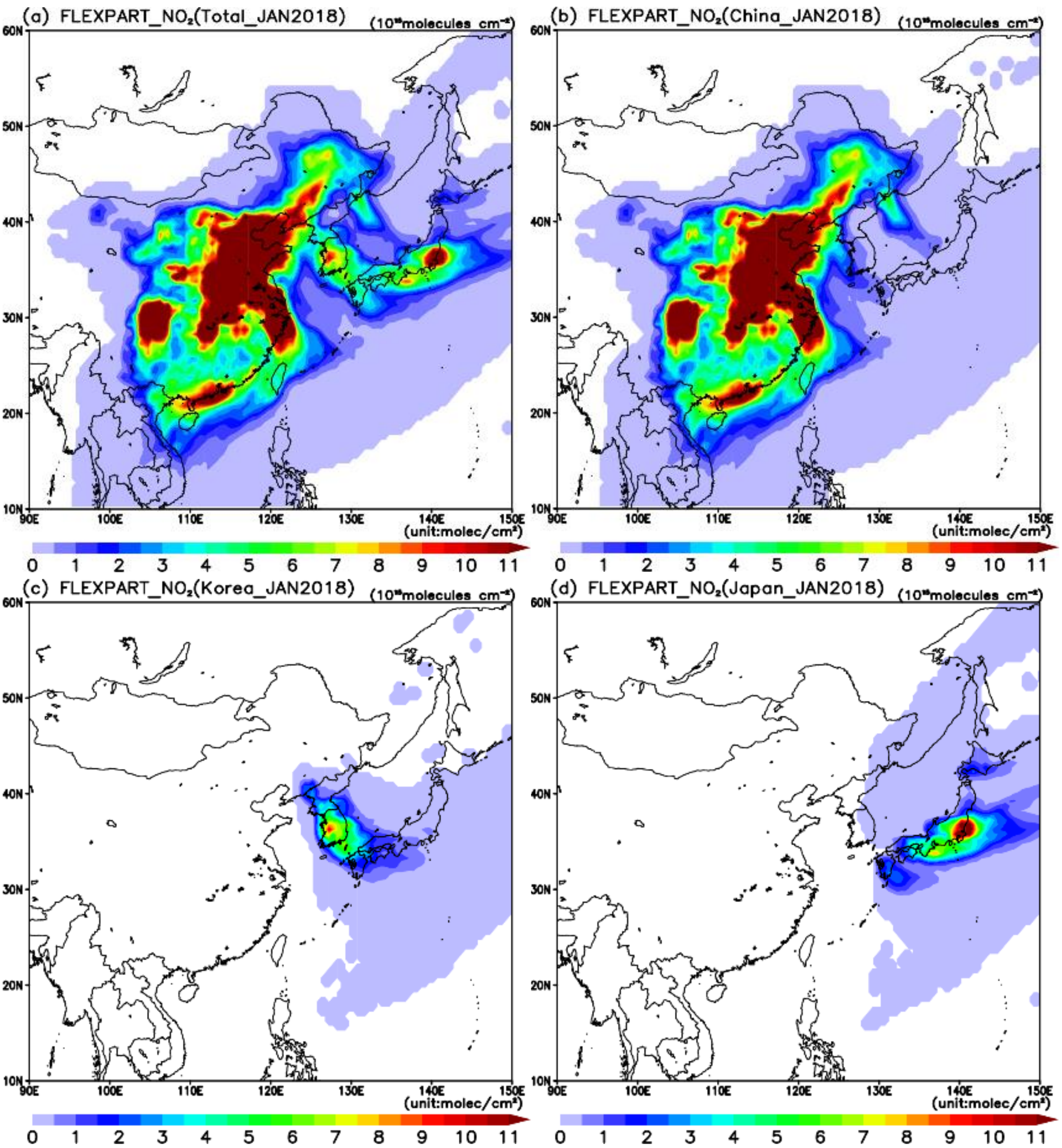


Fig. 3. 2. Spatial distributions of mean NO_2 of FLEXPART simulations separated by sectors indicated in fig. 2. 1. ((a) Total, (b) China, (c) Korean, and (d) Japan)

of emitted NO_2 from each country are southwestward which is following to the wind directions in winter as commonly known.

Model simulation should be appropriately compared to the observations or measurements data due mainly to there are many uncertainty sources in the model simulation. In order to secure the feasibility of model simulation, we firstly validate with OMI tropospheric NO_2 columns as shown in figure 3.3 (left: FLEXPART, right: OMI tropospheric NO_2 measurement). The OMI retrieval has missing pixels, and cover of clouds interrupt providing good quality of spatio-temporal coverage over the regions. Nonetheless, this comparison of main plume locations agrees quite well with the spatial distributions between of them for each event day, even though coarse definitions of the gridded source emissions (2° x 2°).

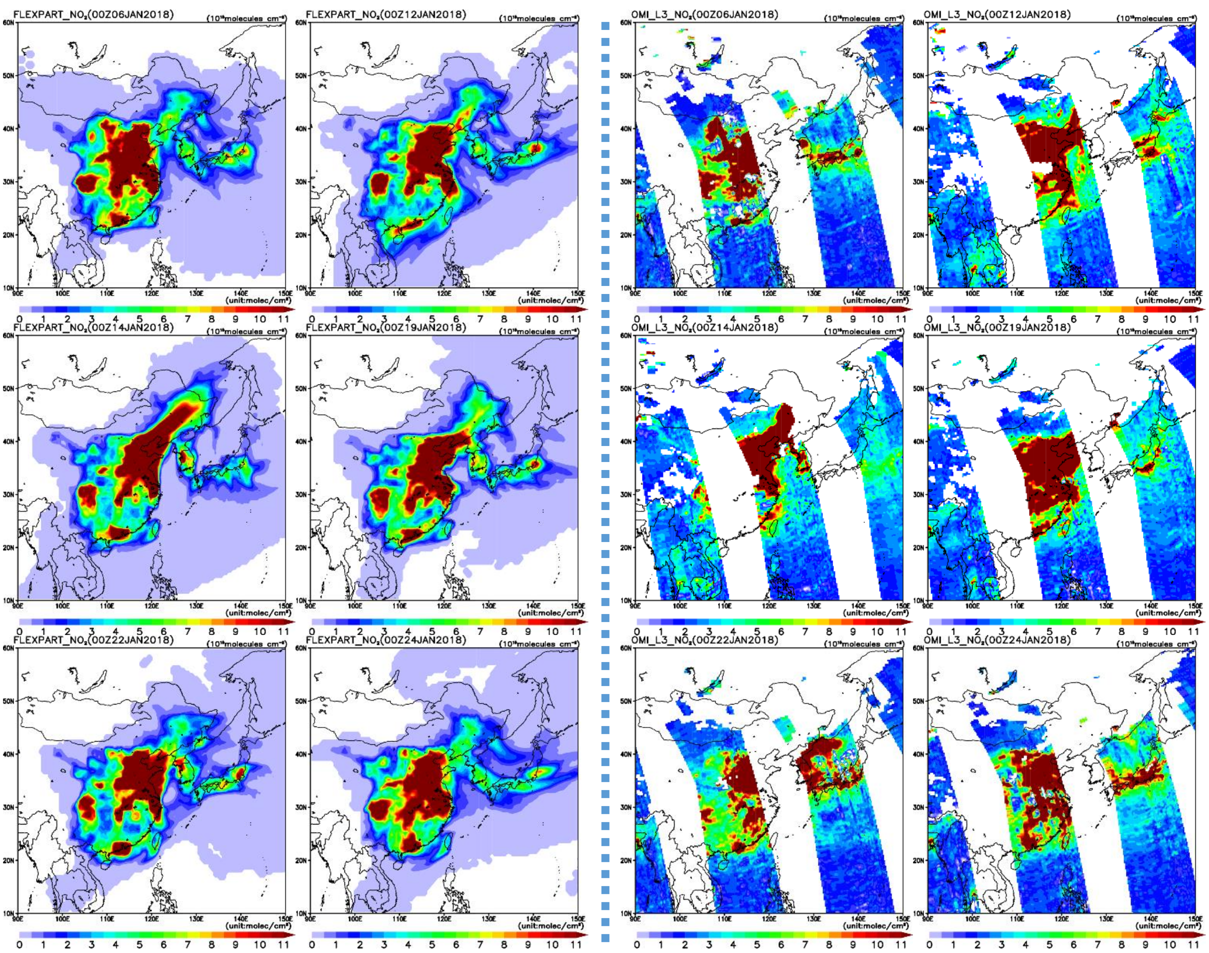


Fig. 3. 3. Comparison of transported NO_2 plume events in January 2018 (FLEXPART (left), OMI (right), January 6, 12, 14, 19, 22, and 24 2018)

Figure 3.4 and 3.5 demonstrate cross-validations of EOF analysis of surface NO_2, OMI tropospheric NO_2 and simulated NO_2 in South Korea. Distributions of OMI and FLEXPART EOF vectors of generally agree among them, which mean that they have pattern correlations over center of the korean peninsula. However, PC time-series do not match in Figure 3.4. Basically, Variability (negative and positive) is not consistent. However, they noticeably have the variability in the areas in which they are affected appears to be consistent.

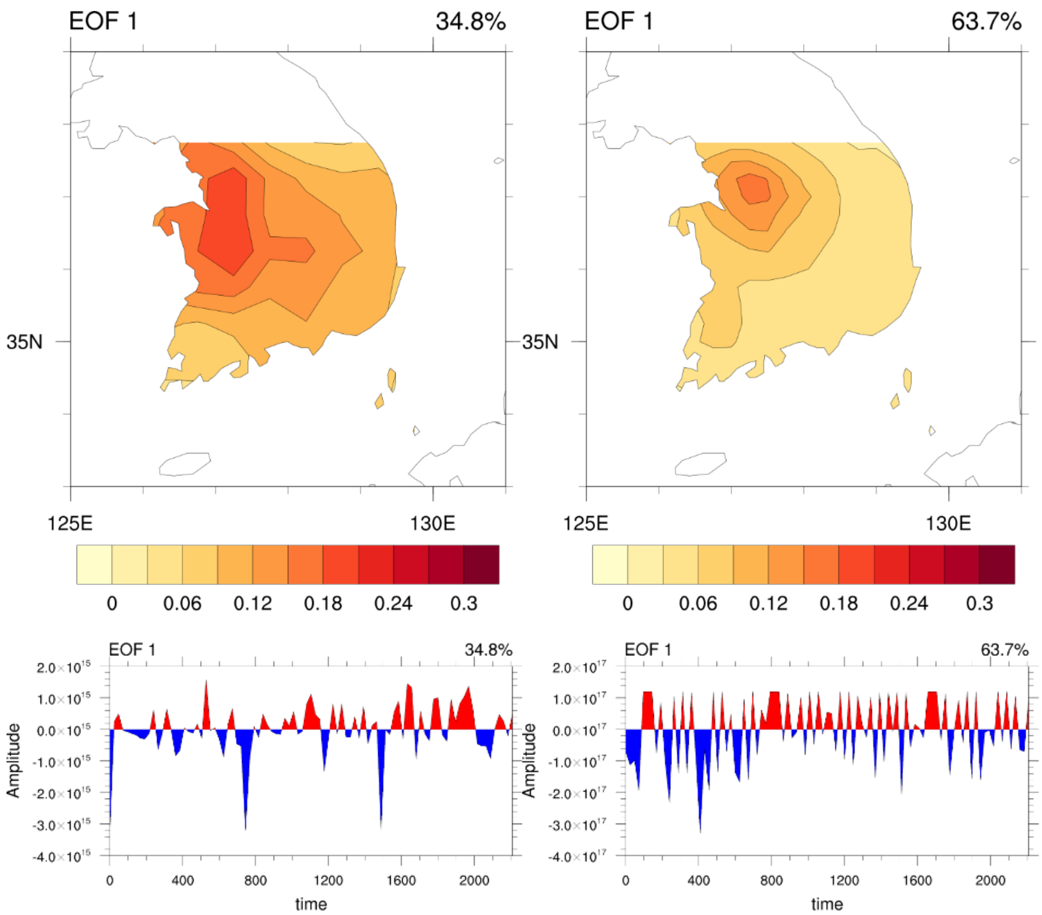


Fig. 3. 4. EOF analysis for simulated year (2011, 2016, and 2018 Jan) in comparison of FLEXPART vs. OMI in unit of column density.

EOF vector shown in figure 3.5 indicates negative variability which shows that the two analysis match. However, the differences in varaibility between Eastern and western side of South Korea occur in models and satellite data, but do not appear conspicuous in surface observations. In addition, PC time-series analysis in figure 3.5 shows well agreement seemingly. As shown in figure 3.6, they have relatively well agreement with high level of correlation (R^2=0.3933)

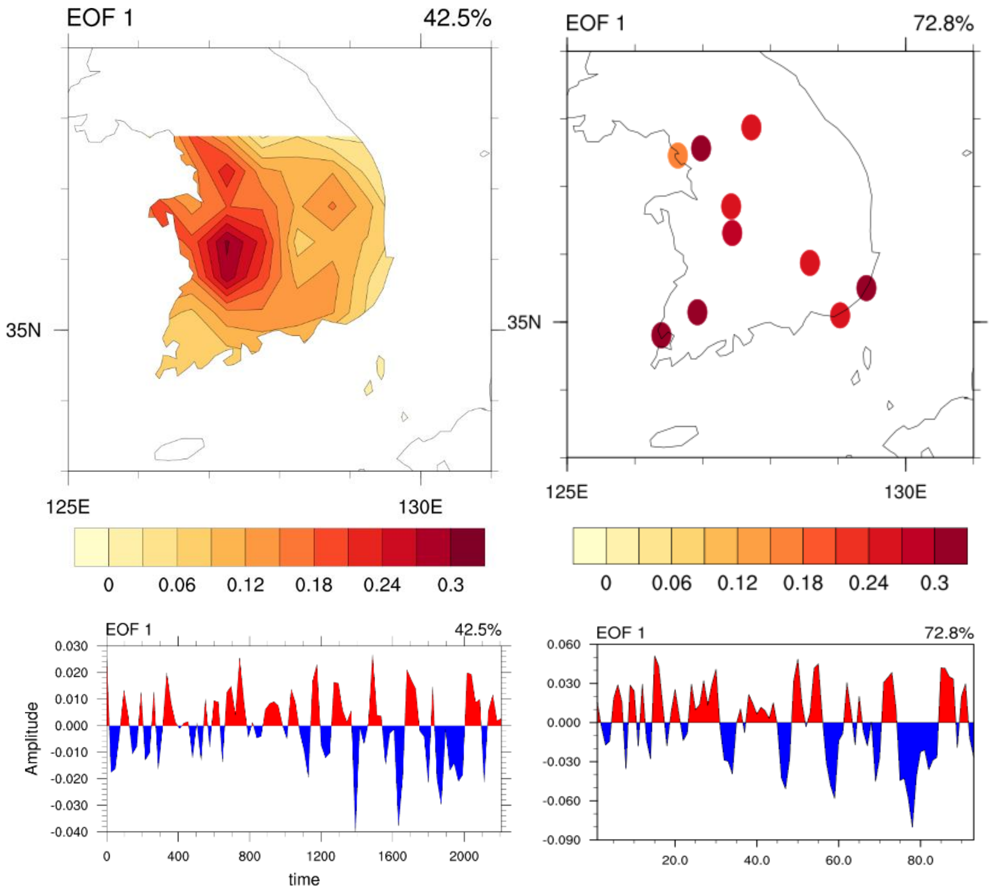


Fig. 3. 5. EOF analysis for simulated year (2011, 2016, and 2018 Jan) in comparison of FLEXPART vs. Surface observations in unit of concentration

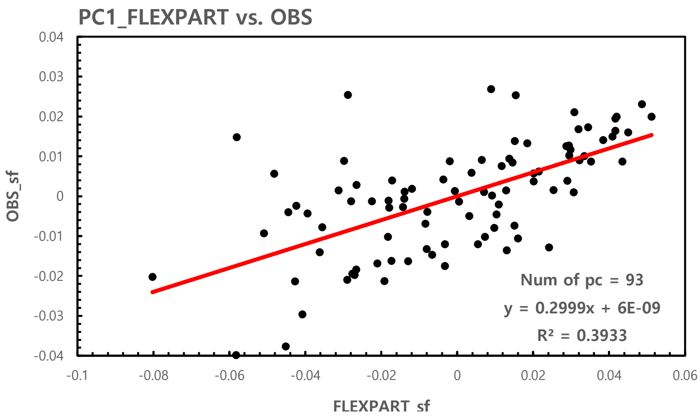


Fig. 3. 6. Scatter plot indicating a correlation of PC time-series between surface observation and FLEXPART.

In fact, However, there remains room for analysis between FLEXPART simulations results and surface observation in different points of view, since there are uncertainty sources which came from gridding emission inventory, the inherent limitations of the model and so on. We will have a more time to discuss about this uncertainty in discussion session later. Consequently, figure 3.5 and 3.6 might imply that this model simulation is not suitable for quantitative evaluations. Taken altogether, FLEXPART simulations are feasible according to comparison of the plume locations validated with OMI retrievals qualitatively, which means it might be suitable for budget analysis preticulary SRRs. In contrast, inter-comparison of surface measurements and OMI retrievals through EOFs are relatively not matched with, which means not suitable for quantitative evaluations.

3.2 Source Receptor Relationships and sensitivity tests

Table 3.1 (figure 3.7) shows the Source-Receptor Relationships (hereafter referred to as SRR) based on sectors we separated as shown in fig. 2.1. The table shown in top demonstrates the summary of the abbreviations used to simplify the SRRs (e.g. Source – China, Receptor – Korea: CK). Middle table and bottom table respectively show SRRs on Jan and DJF for simulations. There are only slight differences among them. We firstly took notice CK where the emissions from China, which account for the largest portion of emissions in East Asia, affect to the Korean peninsula by the dominant downwind. Furthermore, we expected that CK might be gradually declined every year according to the tropospheric NO_2 trend shown in fig. 3.1. No dramatic changes in SRR values were identified in year-by-year, and the NO_2 emitted from china contributing to the Korean peninsula (CK) is about 44-49%. According to previous study, these results are consistent with the low level of contribution shown in the LTP report (45~65%, National Institute of Environmental Research, 2012). NO_2 concentration in China and Japan is mainly predominated by local emission sources. Unlike Japan, China where accounts for a large portion of the total emissions in East Asia and being located on the upwind side significant impacts to the Korean peninsula and Japan. It agrees with our expectation in the experiment on January, however, in the simulation on DJF does not match, rather, the SRR is highest. In this phase, we deduce that there would be other factors contributing SRRs more than total amount of emissions. JJ shows that it has a strong local impact, since the amount of NO_x reached with seasonal winds is small. In summer and fall, the contribution of the locals would be even greater than the winter season, which is predominantly wind driven by the Siberian high with winter monsoon.

First, it is necessary to confirm if these differences are due to emission inventory. We exam simulations with the same meteorological condition (on Jan 2018) with different emission inventories (CREATE2010 and 2015). The result is shown in table 3.2 (figure 3.8). Difference of emission inventories still affect to the migrations of air pollutants but do not cause huge difference which means the long-range transport is dependent on meteorological conditions, especially wind directions and speeds. We also find the significant difference depending on decay-time of simulated NO_2 as shown in Table 3.3 (figure 3.9). Almost all SRRs show logarithmic variations as decay-time changes. However, decay-time over a week is not suitable due to staying for more than a week is unrealistic.

We also conduct experiments of sensitivity test for meteorological conditions in order to confirm which factor is most affectable contributing to SRRs. Table 3.4 (figure 3.9) shows SRRs depending on meteorological conditions with the same emission inventory (CREATE 2015); CREATE 2015 and 2017 are the same amount but not the same of weeks. Simulation on DJF 2017-18 indicates highest value of the entire periods on CK, which mean that emissions have reached the Korean peninsula most through downwind. Also, in this period, CJ and KJ show that downwind transporting emissions is strong enough to reach Japan as shown as highest value in the table 3.4.

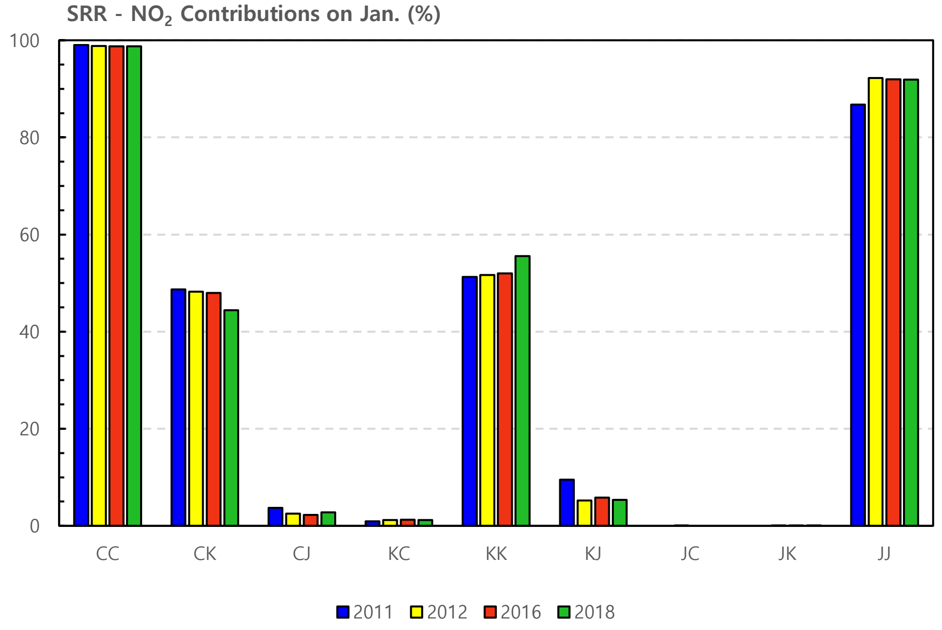
Additionally, we are curious how total amounts of emission contribute to the long-range transport. We set radical changes of total amount of NO_2 on emission inventory and examine the simulations with doubling and halving. Figure 3.7 shows simulations on DJF 2011-10 with radical changes ((a) doubling, (b) control and (c) halving) of emission inventories and control test minus doubling and halving (d) and (e). Absolute concentrations could be changed; however, SRRs scarcely change since the denominators and the numerators increase proportionately. In spite of this reason, there are infinitesimal changes on it since wet scavenging and dry deposition are included in the model simulations which are the non-linearity.

Receptor				
		China	**Korea**	**Japan**
Source	**China**	**CC**	**CK**	**CJ**
	Korea	**KC**	**KK**	**KJ**
	Japan	**JC**	**JK**	**JJ**

JAN	CC	CK	CJ	KC	KK	KJ	JC	JK	JJ
2011	99.05	48.71	3.72	0.95	51.29	9.54	0	0	86.74
2012	98.79	48.24	2.54	1.21	51.67	5.22	0	0.09	92.23
2016	98.75	47.95	2.24	1.25	51.98	5.8	0	0.07	91.96
2018	98.79	44.37	2.77	1.21	55.54	5.32	0	0.09	91.9

DJF	CC	CK	CJ	KC	KK	KJ	JC	JK	JJ
2010_11	99.07	46.27	4.3	0.93	53.61	7.91	0	0.13	87.8
2011_12	98.86	43.74	2.55	1.14	56.24	6.33	0	0.02	91.13
2015_16	98.7	42.79	1.77	1.3	56.99	4.03	0	0.23	94.20
2017_18	98.94	45.98	2.94	1.06	53.96	5.82	0	0.06	91.24

Table 3. 1. SRR table indications of source and receptor regions (top) and SRR on January and DJF in 2011, 2012, 2016 and 2018 (middle and bottom).



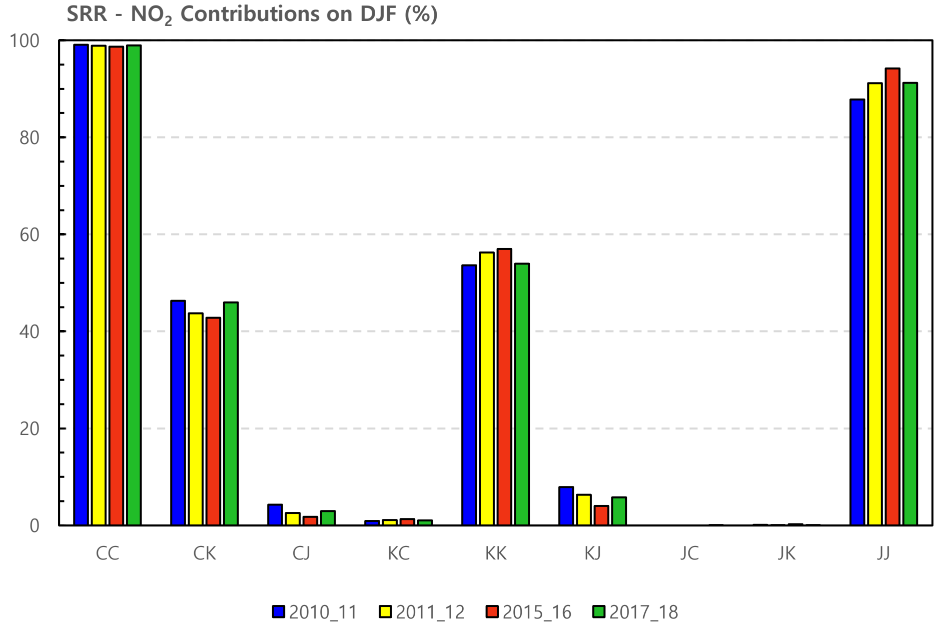


Fig. 3. 7. The bar graphs for table 3. 1 of respective model experiments (Top: January, Bottom: DJF).

EI_2018		Receptor		
Source		China	Korea	Japan
	China	98.79	44.37	2.77
	Korea	1.21	55.54	5.32
	Japan	0	0.09	91.90

EI_2018		Receptor		
Source		China	Korea	Japan
	China	98.9	44.37	2.77
	Korea	1.21	55.54	5.32
	Japan	0	0.09	91.9

Table 3. 2 SRRs of simulation on 2018 JAN with CREATE 2010 (top), and simulation on 2018 JAN with CREATE 2015 (bottom).

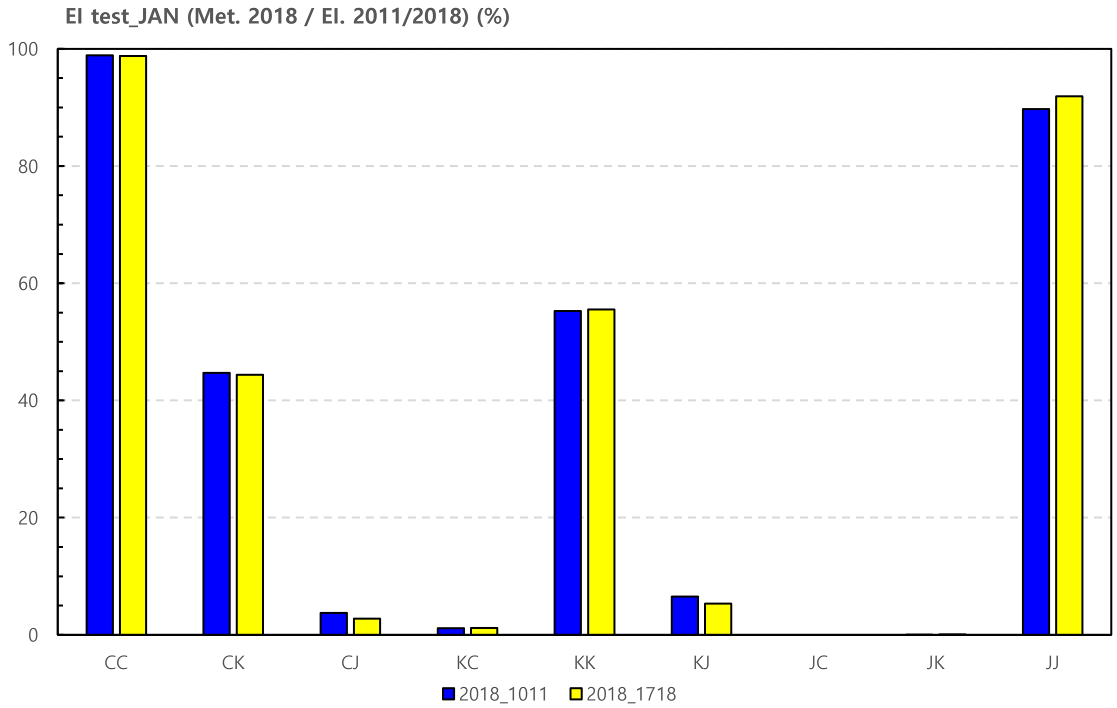


Fig. 3. 8. The bar graphs for table 3. 2 of respective model experiments, which are the same meteorological condition with different emission inventories.

HR	CC	CK	CJ	KC	KK	KJ	JC	JK	JJ
24	98.51	36.05	0.07	1.49	63.92	0.94	0	0.04	99
48	98.66	40.15	0.93	1.34	59.79	3.16	0	0.07	95.91
72	98.79	44.37	2.77	1.21	55.54	5.32	0	0.09	91.9
96	98.87	47.88	5.18	1.13	52.03	6.96	0.001	0.1	87.86
120	98.94	50.55	7.72	1.06	49.34	8.12	0.001	0.11	84.16
144	98.98	52.82	10.40	1.02	47.07	9.2	0.002	0.11	80.4
168	99.02	54.88	12.76	0.98	45.02	9.96	0.003	0.1	77.28

Table 3. 3 SRRs Sensitivity test for decay-time in simulations (from 24-hr to 168-hr)

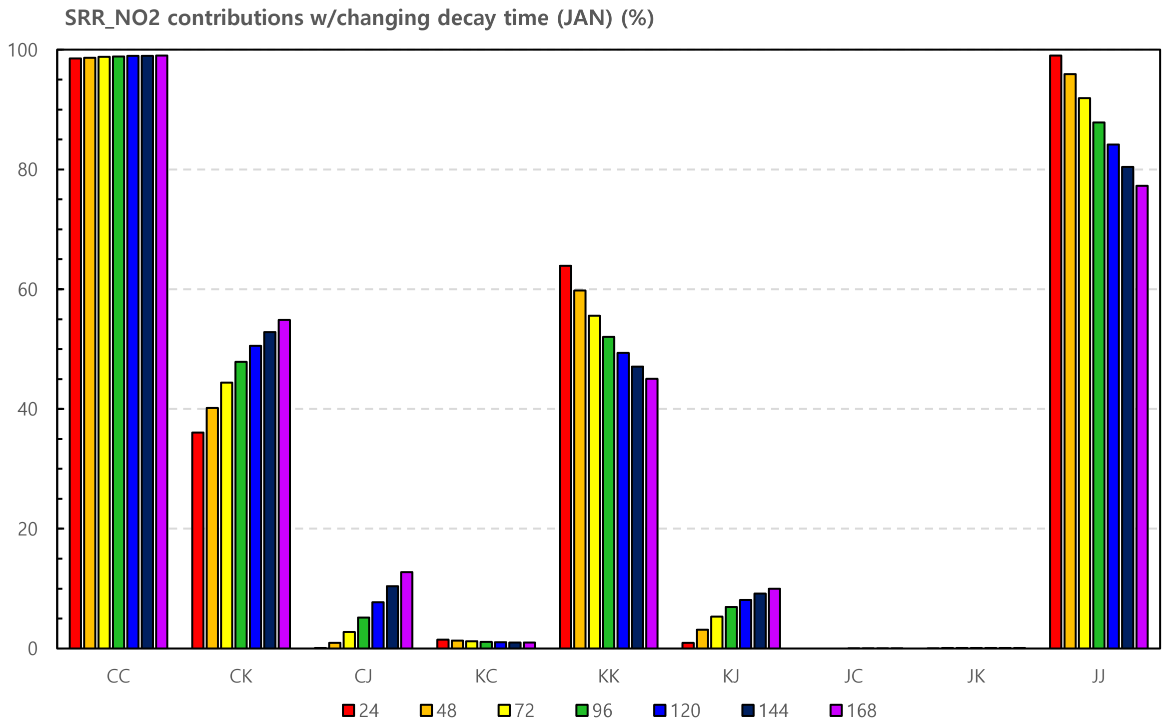


Fig. 3. 9. The bar graphs for table 3. 3 of respective model experiments, which are the same meteorological condition and emission inventories with different decay times ranging from 24-hr to 168-hr).

2010-11/EI_2015		Receptor		
		China	**Korea**	**Japan**
Source	**China**	99.08	48.14	3.33
	Korea	0.92	51.69	6.65
	Japan	0	0.18	90.02

2010-11/EI_2015		Receptor		
		China	**Korea**	**Japan**
Source	**China**	98.7	42.79	1.73
	Korea	1.3	56.99	4.03
	Japan	0	0.23	94.2

2010-11/EI_2015		Receptor		
		China	**Korea**	**Japan**
Source	**China**	98.94	45.98	2.94
	Korea	1.06	53.96	5.82
	Japan	0	0.06	91.24

Table 3. 4 SRRs sensitivity test depending on meteorological conditions (2010-11, 2015-16 and 2017-18 DJF) with CREATE 2015 and CREATE 2017.

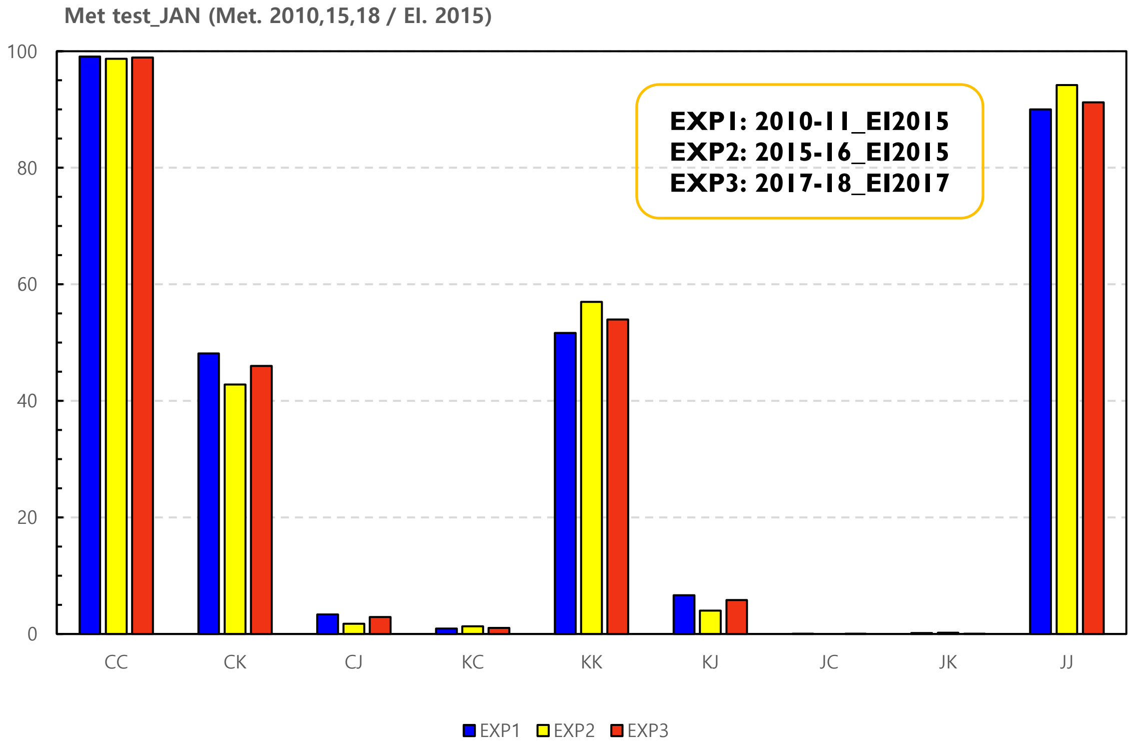


Fig. 3. 10. The bar graphs for table 3. 4 of respective model experiments, which are the same emission inventory with different meteorological conditions.

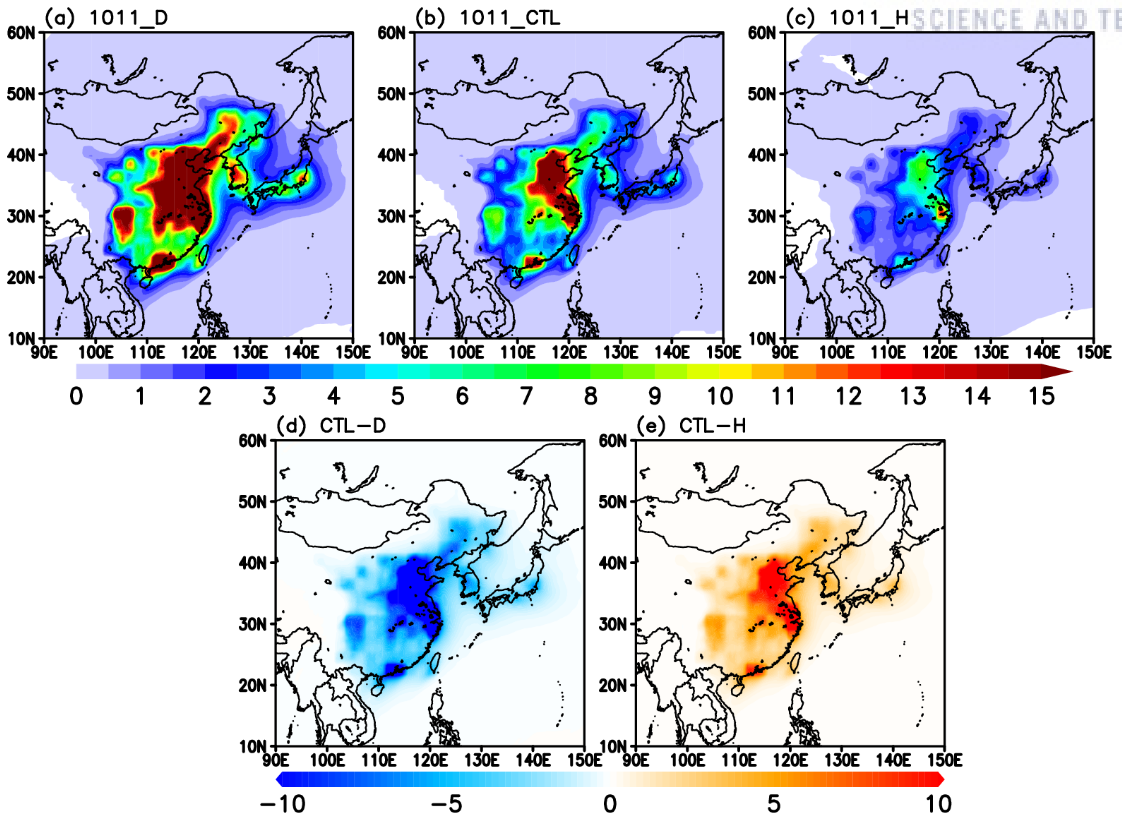


Fig. 3. 11. Evaluations of radical changes of emission inventories ((a) doubling, (b) control, (c) halving, and control test minus doubling and halving test (d) and (e)).

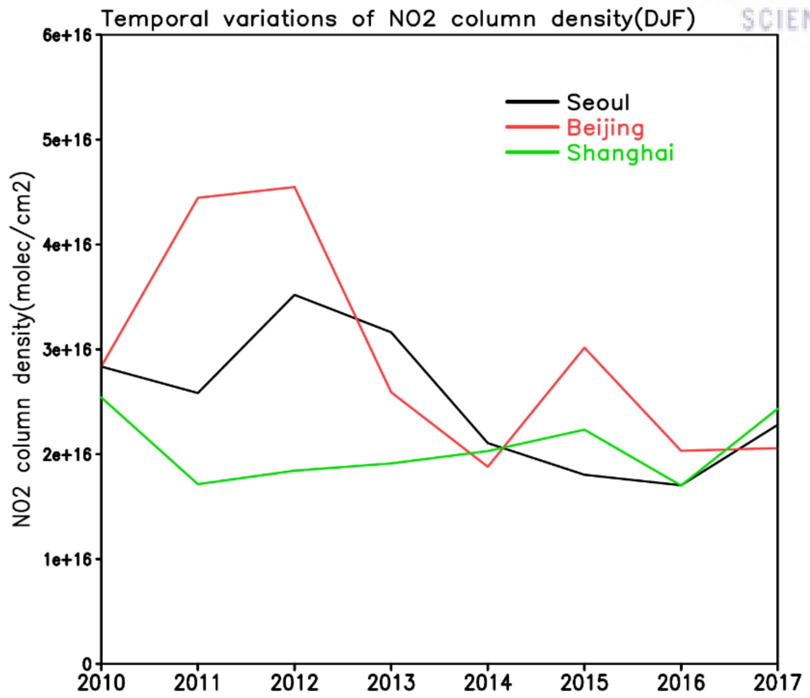
		2010_DJF_D (1011_D)			2010_DJF (CTL)			2010_DJF_H (1011_H)		
		Receptor			**Receptor**			**Receptor**		
		China	Korea	Japan	China	Korea	Japan	China	Korea	Japan
Source	China	99.09	46.14	4.31	99.07	46.27	4.3	99.08	46.35	4.27
	Korea	0.91	53.74	8.08	0.93	53.61	7.91	0.92	53.54	7.86
	Japan	0	0.12	87.61	0	0.12	87.79	0	0.11	87.87

Table 3. 5 Evaluations of radical changes of emission inventories through SRRs (red-colored column: doubling, blue-colored column: control and yellow-colored: halving)

Chapter IV

Discussion

Back to the beginning, we can find a similar pattern when the comparing the time-series of the major cities in East Asia with the simulated NO_2 column density as shown in fig. 4.1. We assume that it is because the total amount of emissions base year of 2015 has been reduced compared to the emissions base year of 2010 in CREATE inventories. Thus, it can plainly explain the sensitivity of emission inventories. However, unlike OMI retrievals, the column density in Shanghai is larger than Beijing and Seoul. As briefly mentioned above, it is due to the gridding process causing the amount of emission is not quantitatively and accurately matched in that location. Uncertainty sources are hidden while the emission is gridded into 2° **x** 2°. Plus, it also can cause emission shift and restricting characteristics of emission sources. As widely known, activity data used in integrating anthropogenic emissions consists of point, line and are, and the intrinsic characters contained in each emission information is corrupted during the gridding process which increases error bias. For instance, industrial areas and power plants which are the strong contribution emission sources to anthropogenic emissions. Power plants one representative of point source air pollutants, however, once it is processed of gridding, they are limited on expressing a steeply increase of emissions from point sources. It was explained by Lee et al., 2014 that spatially shifts in the locations of the simulated plumes due to the coarse resolution of the source regions. Through this, it can be consequently explained that the limitations of quantitative evaluations of EOF analysis in this experiment. The inconsistency of the EOF analysis on the Korean peninsula can be attributed to degradation of emission characteristics caused by the gridization process. Emission inventory gridded by 2° **x** 2° is too causal to say that it reflected precise and detailed information about emissions on the Korean peninsula. Moreover, since the quantitative evaluation of the transport model is strictly limited, it is more appropriate to identify the long-range and trans-boundary transport mechanism through a qualitative evaluation, budget analysis especially SRRs, rather than quantitative evaluation.



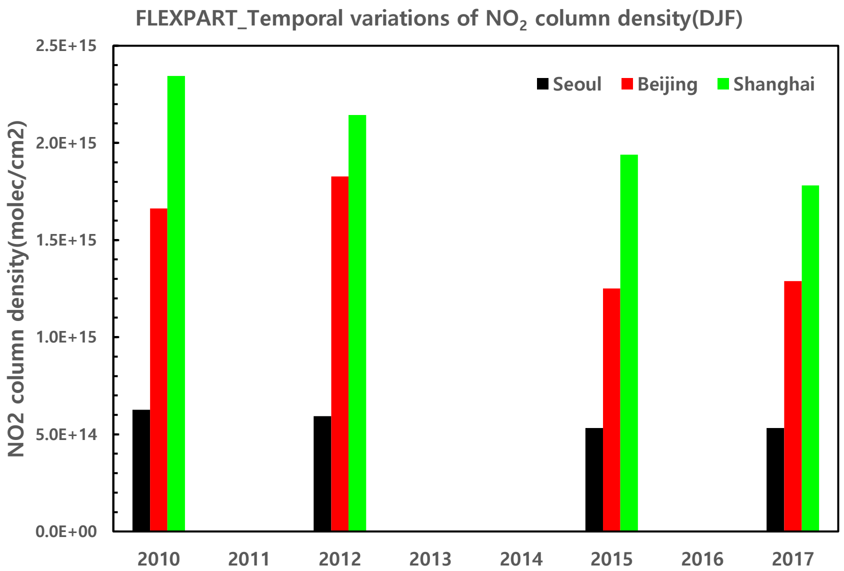


Fig. 4. 1. Yearly temporal variations of OMI tropospheric NO_2 column density (DJF) over major city in Korea and China (top), and FLEXPART NO_2 column density (DJF, down).

According to the simulation results and SRR analysis, the unexpected increasing of CK value, which occurred on DJF 2018, was due to neither given meteorological conditions and seasonality nor total amounts of emissions. To investigate synoptic scale meteorological conditions in detail, wind directions and speeds at 850-hpa are drawn to compare synoptic patterns of wind vectors for the respective experimental period as shown in fig. 4.2. During winter season, a continental anticyclone placed adjacent China is usual, and strong northerly and northwesterly winds lead to long-range transport with great dilution compared to other seasons (Lee et al., 2014). The wind data is plotted with CFSv2 which is employed in the model simulations. Wind directions and speeds are relatively faster than other periods as indicated as blue circled, however, this clue are still not enough to explain the unexpected CK value in 2018 since it is seemingly not significant differences affecting to the long-range transport. According to Kim et al., (2017b), they found and addressed that there is high correlation between variations of surface pollutants and wind speed. Following to this study, we confirm the inverse relationship between wind speeds and air pollutants in the lower level. Fig. 4.3 shows anomalies of DJF mean 10m-winds (WSPD10) of CFSv2 meteorological input data (blue) and anomalies of simulated NO_2 column (yellow) over South Korea. The noticeable fact indicated in the figure is that its inverse proportion between of them is obvious which also shows R^2=0.864. Mean 10m-wind speeds over south in 2017-18 DJF is the slowest compared to other periods, so that SRR on DJF 2018 reaches to the highest value.

Except for experiments considering this synoptic scale of meteorological conditions, the biggest factor influencing on SRRs is the decay-time changes. Usually, the life-time of NO_2 is at least ~1 day in boundary layer. In this experiment, we determine the decay-time 72-hr longer than usual, in order to overcome the absence of chemical conversion (including re-conversion process) and considering that the model is an off-line model. We also extend decay-time ranging from 24-hr to 168-hr, and we analyze how the changes of durations of NO_x are influence to the SRRs. As mentioned above, CK increase logarithmically, and other SRRs show logarithmical changes depending on decay-time as well. This suggests that seasonal variations that determine that decay-time of NO_2 may have a large impact on long-range transport; In summer, photochemical reaction rates are peak (~6-hr), so decay time of NOx is relatively reduced, moreover, north and northeast winds become dominant, the emissions from China transport to northern part of China and accumulate. In contrast, anthropogenic NO_x emissions from China do not change significantly with season (Zhang et al., 2007) which means the changes in the contribution of transported NO_2 caused by seasonal changes implies dependence on meteorological conditions.

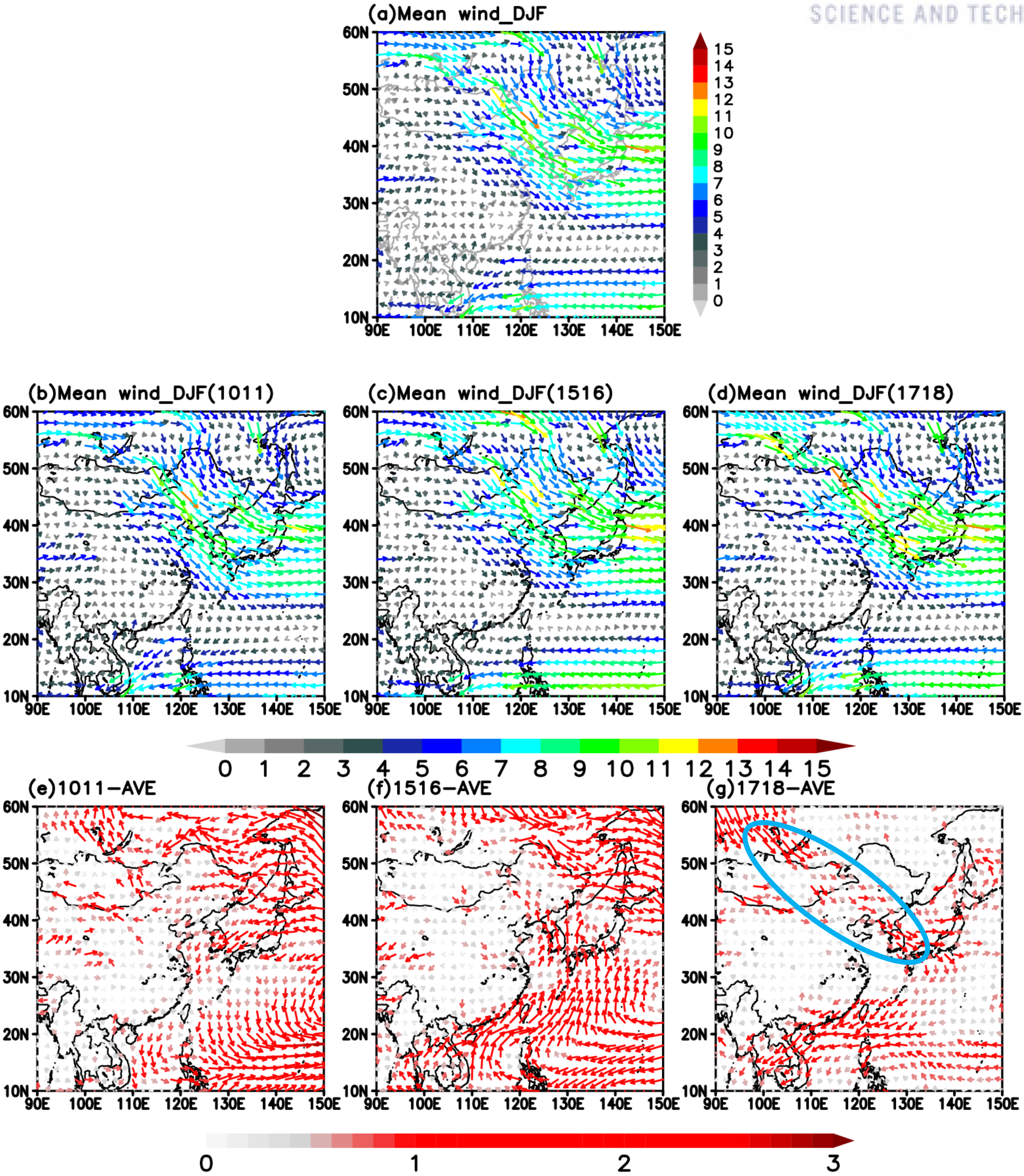


Fig. 4. 2 DJF mean wind directions of CFSv2(1011, 1516, and 1718) at 850hpa. (b), (c) and (d) are DJF mean wind directions for each year. (e), (f) and (g) are differences between each year and averaged value.

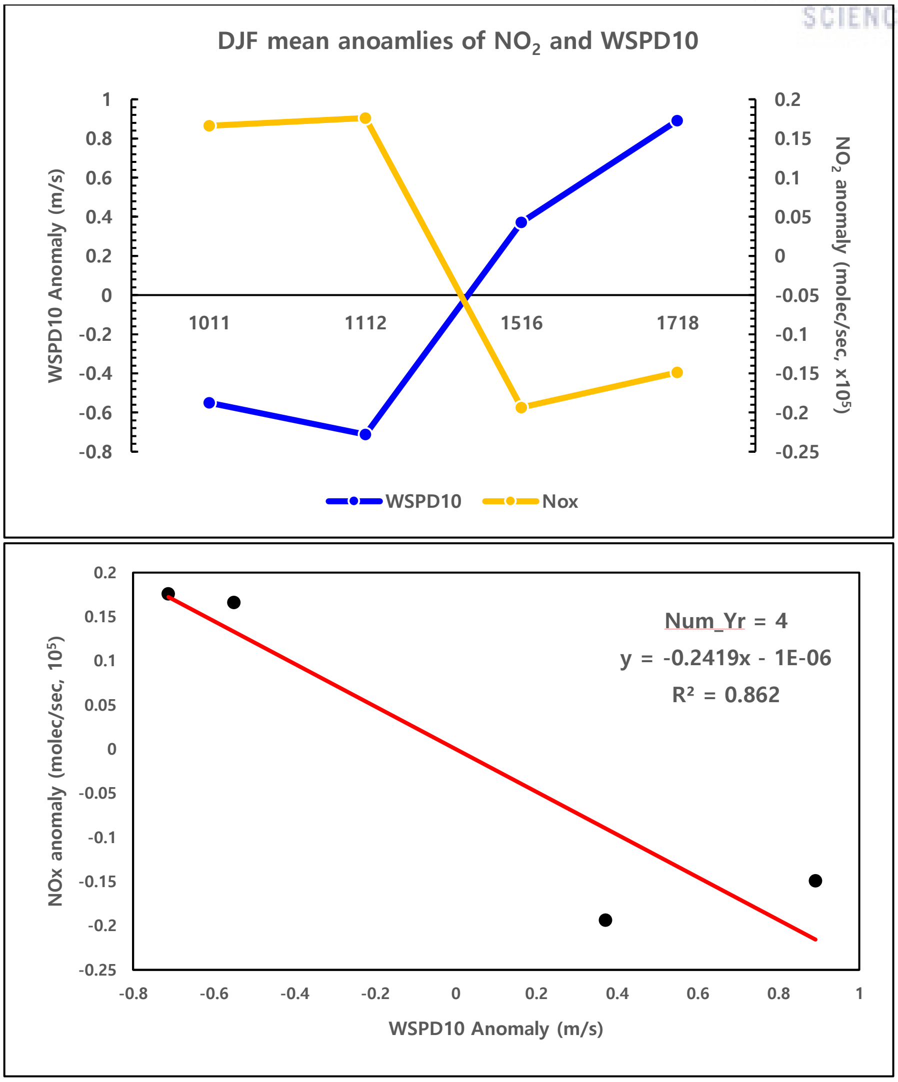


Fig. 4. 3. (top) Anomalies of DJF mean 10m-winds (CFSv2, blue line) and NO_x column density simulated by FLEXPART (Yellow line) during simulation periods (2011, 2012, 2015 and 2017 DJF), and (down) scatter plot between anomalies of WDPS10 and NO_x.

Chapter V

Summary and Concluding Remarks

This study analyzes variations of the pollutants, NO_2, in the winter season, using the multi-year model simulations, and investigate the mechanism of long-range and trans-boundary transport. The results of various previous studies show that the impact of the seasonal variations is largely influenced to the atmospheric transport of air pollutants. In this experiment, we integrate the factors affecting to the long-range transport, which are meteorological conditions, emissions, and chemical conversion (altered to decay-time) and reflect into multi-year simulations. This study also supports the previous study that transported and emitted NO_x are both important for better understanding the local NO_x emissions over the East Asia (Lee et al., 2014). Since 2010, emissions in East Asia has been generally decreased especially China, however, SRRs in Korea and Japan are not much affected because meteorological conditions are not ignorable factors, which mean that the impact of the local emissions are increased.

Although emission control efforts from the South Korean government and communities, as well as neighboring countries, recent changes of meteorological conditions around the East Asia seem to counteract those efforts (Kim et al., 2017b). As mentioned in the discussion section, it is estimated that the relationship between wind speeds and pollutants due to seasonal variability has the greatest contribution to SRR analysis in terms of synoptic scale of meteorological conditions. In conclusion, we found that the contribution of the estimated factors is not only large portions of SRRs, but it could be a determinant of contribution. It was also confirmed that decay-time changes, which had the second most significant impact, were the important factor for long-range transport due to seasonally varying. In addition, long-range and trans-boundary transport occurs due to the complex interactions of the factors mentioned above, and the factors affecting the stay are the transport of the receptor area, regional scale of meteorological conditions, wind directions and speeds.

To sum up, current situations of air quality we faced, especially atmospheric long-range transport are a combination of long-term decline by various attempts of reducing emissions and variations of synoptic scale of meteorological conditions especially wind directions and speeds. In order to get better understanding of this complex process, it is worth to sustained investigations of long-term and multivariate analysis.

Chapter VI

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더불어 바쁘신 와중에도 학위 논문 심사 간 위원으로서 진심 어린 조언으로 길라잡이가 되어 주신 송창근 교수님과 최성득 교수님께도 감사의 말씀 올립니다. 사실 비전공자로서 익숙치 않은데 다가 더불어 심도 깊은 대학원 강의에서 지치기 일쑤였음에도 늘 가슴 벅차오르는 강의를 선물해주신 송창근 교수님, 두서없이 찾아 뵈어도 늘 따뜻한 웃음으로 반겨 주시며 날카로운 지적으로 연구에 모자람이 없게끔 많은 지도 편달 주신 최성득 교수님, 앞으로도 멋진 모습 변치 않으시고 늘 밝게 빛나는 모습으로 계시길 기원합니다.

제 학위 논문이 진행될 수 있게끔 큰 발판을 마련해주신 주신 부산대학교의 이효정 박사님, 그리고 발돋움을 할 수 있게 다방면으로 도움을 주신 김현국 박사님. 바쁘신 와중에도 본인의 우문에 늘 현답으로 지표를 던져 주신 두 분의 박사님들 덕분에 부족한 제 학위 논문이 보다 값지게 되지 않았나 싶습니다. 이에 감사의 말씀 올리고 싶습니다.

길다면 길고 짧다면 짧은 기간 동안 UCEM 연구실 여러분들께도 신세를 많이 졌습니다. 시작부터 많은 도움 주시고 지금은 미국에서 열정적으로 본인의 삶을 개척해 나가고 계시는 김동민 박사님, 이에 뒤따라 늘 스스로를 돕는 성윤이형 두 분 모두 진심으로 행복하셨으면 좋겠습니다. 연구실의 맏언니로 듬직하지만 편안한 혜림이, 자신과 동시에 주변사람도 챙길 줄 아는, 이제는 강대현 박사님, 이 곳에 자리 잡을 수 있게끔 출발선에 서기 전부터 많은 도움을 준 은교, 같은 분야를 공부하며 한계에 부딪힐 때마다 그 벽을 넘을 수 있게 많은 도움 준 강한이, 남은 연구실 생활 슬기롭게 잘 마치고 본인들의 삶에 늘 행복이 가득했으면 좋겠습니다. 지난 1 년 간 랩장으로서 궂은 일 도맡아 솔선수범하며 보이는 곳은 물론 보이지 않는 곳에서까지 많이 챙겨준 낙빈이, 늘 긍정적인 에너지로 주변에 활력을 불어넣어주는 승희, 운동이면 운동 공부면 공부 못하는 게 없는 늘 밝은 민상이, 그리고 행복한 아틀란티스 소녀 같이 밝은 에너지로 부족한 저를 많이 챙겨준 선주, 너무 고맙습니다. 이제 들어올 선래와 지해에게는 제가 미쳐 보지 못했던, 하지 못했던 많은 부족한 것들을 반면교사 삼아 부디 본인들의 길을 지혜롭게 헤쳐 나아가길 기원하며 지금처럼 늘 웃는 모습 간직했으면 좋겠습니다. 그리고 UCEM 을 거쳐간 언급하지 못한 학부생 인턴 여러분들도 짧게나마 고마움을 전합니다. 이렇게 한 줄로 짧게 요약하기엔 너무나도 대단하고 고마운 UCEM, 이곳에서 당신들을 만난 건 정말 큰 행운이었습니다. 정말 고맙습니다.

비록 같은 연구실은 아니었지만, 정신적으로 큰 버팀목이 되어준 CDL 연구실 친구들. 본인의 행복 찾아 길을 나선 뒤, 지금은 너무나도 행복한 박기웅 소위님, 때로는 철부지처럼 때로는 큰 어른처럼 정말 배울 점이 많은 진짜배기 예철 "갓", 천진난만한 웃음으로 힘들 때 웃는 게 일류라는 걸 스스로 증명해 나가고 있는 도연이, 정말 친동생처럼 잘 따라주고 많은 아낌없는 응원을 준 한준이 아프지 말고 늘 행복하길 기원합니다. 그리고 잘 알면서 모르지영 진짜 (골골)아프지 말고 열심히 해서 전우님과 같이 졸업하기를 바라며, 찬영이도 미국가서도 지금처럼 열심히 아니 지금보다 더 정진하여 본인들이 뜻한 바 모두 이룩하기를 기원합니다. 지금은 지스트에서 열심히 성장하고 있는 깐돌이 기연이, 파티션 너머로 보이지 않게 든든한 우군이 되어준 벤큡 동기 서희, 어디를 가서든 본인이 가진 역량 그 이상으로 해낼 거라 믿어 의심치 않습니다. 새로운 환경에서 이제 성공적인 첫 발을 디딘 있는 은령씨, 그 언니와 동생 같은 예솔이, 에어랩의 차세대 신형 엔진 강호, 서서히 조금씩, 하지만 견고히 초석을 다져 나가며 그대들 만의 에어랩을 멋지게 꾸려 나가길 기원합니다. 더불어 제가 학업 및 연구에 전념할 수 있게끔 다방면으로 도움을 주신 행정실 선생님 여러분들께도 감사의 말씀 전합니다.

스무 살의 초중반을 함께한 대학 동기들, 강철 토목 A 반 끝까지 응원해주고 격려해줘서 너무 고맙습니다. 다들 무사히 자리잡고 안정을 찾아가는 모습을 보니 너무 행복합니다. 멀리 떨어진 곳에서도 가끔 힘들고 지칠 때 이해해주고 공감해주신 WDPL 의 형누나들 너무 감사합니다. 호주로부터 온 새해인사, 이제서야 답장합니다 고마워 유진아 먼 곳에서 아프지 말고 늘 행복했으면 좋겠습니다. 제 인생 다섯 손가락 안에 머물러 계시는 이오쌤, 제 스스로의 가능성에 의문이 생길 때 마다 독려의 말 한마디로 망설임을 꺾어 주셔서 정말 감사합니다. U-SURF 부터 룸메 착한 권형이 아프지 말고 늘 건강하고 밝은 모습으로 무사히 졸업하길 바랍니다. 그리고 인생 최고의 친구들 'MMF', 오민섭, 원종현, 장성호, 장재봉 그리고 한종현 (가나다 순). 나중에 노인정에서 음악 볼륨 최대로 지팡이 짚고 춤추고 놀아야 되니까 그때까지 한 명이라도 아프지 말고 건강하고 늘 행복하자^^. 이 외에도 너무 많은 분들에게 신세를 졌습니다. 한 분 한 분들께 직접 말씀드릴 순 없지만 기회가 닿는다면, 아니 기회가 닿지 않더라도 마음을 다해 감사의 말씀을 꼭 전하고 싶습니다.

본인이 힘들고 지칠 때 멀지만 가장 가까운 곳에서 진심 어린 응원으로 묵묵히 지켜봐 준 김민지. 본인의 석사과정이라는 이야기에 훌륭히 마침표를 찍을 수 있게 해준 가장 큰 원동력이 되어 주었습니다. 후회될 일 없게, 그리고 또 후회하지 않게 꼭 고마움과 함께 진심이 담긴 사랑을 전합니다. 이제야 출발선에 선 우리, 행복한 이인삼각이 될 수 있게 끊임없이 노력하겠습니다. 본격적으로 20 대에 접어 든 후, 계속 집 밖을 배회하느라 집안에 많이 무심했던 못난 형을 둔 내 동생 봉재. 부족한 형을 대신해 가족들을 챙겨주어서 너무 고맙다. 대학원 진학이라는 쉽지 않은 결정을 지지해 주시고 또 제 앞날을 응원해주고 계신 가족 친지 여러분들께 감사의 말씀을 올립니다. 바쁘고 정신없었다는 핑계 뒤에 숨어 사랑한다는 말 한마디 끝내 전해드리지 못한 할머니. 아프지 않은 하늘에서도 못난 손주 응원해주고 계시겠지요. 사랑합니다.

끝으로, 아직도 철부지에 못미더운 아들 묵묵히 지켜 봐주시고 또 응원해주신 부모님. 당신들의 무한한 지지가 있었기에 지금 이곳에 제가 서 있을 수 있습니다. 사랑하는 부모님께 그리고 제가 사랑하는 모든 분들께 제 작은 결실로 보답하고자 합니다.

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